Quantum diffusion of dipole-oriented indirect excitons in coupled quantum wells

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(May 06, 2002)

A model for diffusion of statistically-degenerate excitons in (coupled) quantum wells is proposed and analysed. Within a microscopic approach, we derive a quantum diffusion equation, calculate and estimate the self-diffusion coefficient for excitons in quantum wells and derive a modified Einstein relation adapted to statistically-degenerated quasi-two-dimensional bosons. It is also shown that the dipole-dipole interaction of indirect excitons effectively screens long-range-correlated disorder in quantum wells. Numerical calculations are given for indirect excitons in GaAs/AlGaAs coupled quantum wells.

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I. INTRODUCTION

A system of indirect excitons in GaAs/AlGaAs coupled quantum wells (QWs) is a unique object for studing the transport and collective properties of interacting quasi-two-dimensional (quasi-2D) composite bosons. This is due to (i) a well-defined dipole-dipole repulsive interaction between indirect excitons, which is not sensitive to the internal spin structure of the particles (the exchange interaction is extremely weak so that the degeneracy factor $g = 4$), (ii) the absence of quasi-2D excitonic molecules, (iii) strong suppression of the interface polariton effect and a long radiative livetime of excitons, and, as we show below, (iv) effective screening of in-plane QW disorder by interacting indirect excitons.

The indirect excitons in coupled GaAs/AlGaAs have been extensively studied in various optical experiments (see, *e.g.*, [1–6]). In particular, a specific jump of the signal intensity in photoluminescence (PL) dynamics of indirect excitons and its nonlinear behaviour have been interpreted in terms of Bose-Einstein (BE) stimulated scattering of statistically-degenerate Bose-particles [2]. Furthermore, very recently a substantial progress has been achived in spatial confinement of quantum-degenerate indirect excitons by using a dip potential trap naturally-grown in GaAs/AlGaAs coupled QWs [3]. A theory of the acoustic-phonon-assisted relaxation kinetics of statistically-degenerate QW excitons has been developed in [7–9].

For a dilute (quasi-) equilibrium gas of indirect excitons the degeneracy temperature T_0 and the chemical potential μ are given by

$$T_0 = \frac{\pi}{2}\left(\frac{\hbar^2}{M_{\rm x}}\right) n_{\rm 2d} \quad \text{and} \quad \mu = k_{\rm B} T \ln\left(1 - e^{-T_0/T}\right) , \tag{1}$$

where $M_{\rm x}$, $n_{\rm 2d}$ and T are the in-plane translational mass, concentration and temperature of excitons, respectively. Crossover from classical to quantum, BE statistics occurs at $T \sim T_0$. The aim of the work is to develop a theoretical model for diffusive in-plane propagation of statistically-degenerate QW excitons.

II. OUTLINE OF THE QUANTUM DIFFUSION THEORY

We assume that exciton-exciton interaction is dominant ($n_{\rm 2d} \geq 10^9\,{\rm cm}^{-2}$, according to the estimate given in [7]) so that a system of quasi-2D excitons can be described in terms of the local quasi-equilibrium (thermodynamic) states. The self-diffusion flux density of QW excitons, $\mathbf{J}_{\rm x}^{\rm diff}$, is given by

$$\mathbf{J}_{\rm x}^{\rm diff} = -D_{\rm x}^{(2d)} \nabla n_{\rm 2d} - \frac{D_{\rm T}^{(2d)}}{T} \nabla T \,, \quad \text{where}$$
$$D_{\rm T}^{(2d)} = \frac{T}{2}\left[n_{\rm 2d} l_{\rm 2d} \frac{\partial v_{\rm 2d}}{\partial T} - n_{\rm 2d} v_{\rm 2d} \frac{\partial l_{\rm 2d}}{\partial T} \right] \quad \text{and}$$
$$D_{\rm x}^{(2d)} = \frac{1}{2}\left[n_{\rm 2d} l_{\rm 2d} \frac{\partial v_{\rm 2d}}{\partial n_{\rm 2d}} + l_{\rm 2d} v_{\rm 2d} - n_{\rm 2d} v_{\rm 2d} \frac{\partial l_{\rm 2d}}{\partial n_{\rm 2d}} \right] . \tag{2}$$

Here, $D_{\rm x}^{(2d)}$ and $D_{\rm T}^{(2d)}$ are the diffusion and thermal diffusion coefficients, respectively, $l_{\rm 2d}$ is the mean free path of excitons and $v_{\rm 2d}$ is their average thermal velocity.

The mean free path of QW excitons is determined by $l_{\rm 2d} = v_{\rm 2d} \tau_{\rm x-x}^{(2d)}$, where $\tau_{\rm x-x}^{(2d)}$ is a characteristic scattering time due to exciton-exciton interaction. Thus one derives

$$l_{\rm 2d} = v_{\rm 2d} \frac{2\pi C \hbar^3}{|u_0|^2 n_{\rm 2d} M_{\rm x}} \,, \tag{3}$$

where C is a numerical constant of the order of unity, u_0/S is the potential of exciton-exciton interaction and S is the QW area.

For small transferred momenta in exciton-exciton scattering, u_0 is given by

$$u_0 = \pi \frac{\hbar^2}{\mu_{\rm x} \chi(d)} \,, \tag{4}$$

where $\mu_{\rm x}$ is the reduced exciton mass and d is the effective separation between the electron and hole layers. The

dimensionless function $\chi(d)$ has two well-defined limits, at $d \ll a_{2d}^{B}$ and $d \geq a_{2d}^{B}$ [$a_{2d}^{B} = (\hbar^2 \varepsilon_b)/(2\mu_x)$ is the 2D exciton Bohr radius and ε_b is the dielectric constant]. The first limit corresponds to the exciton-exciton exchange interaction in single QWs. In this case $\chi(d=0)$ is very close to unity [7], i.e., $\chi(0) = 1.036$, according to first-principle calculations [10]. The second limit deals with a well-defined dipole-dipole interaction of indirect excitons in coupled QWs. In this case $\chi(d \geq a_{2d}^{B}) = a_{2d}^{B}/(2d)$ and Eq. (4) reduces to $u_0 = 4\pi(e^2/\varepsilon_b)d$. The latter is consistent with the plate capacitor formula. Thus we approximate

$$\chi(d) = \begin{cases} 1\,, & d \ll a_{2d}^{B} \\ a_{2d}^{B}/(2d)\,, & d \geq a_{2d}^{B}\,. \end{cases} \tag{5}$$

Using Eqs. (3)-(4) one gets from Eq. (2):

$$D_x^{(2d)} = D_x^{(2d)}(T, T_0) = C\frac{\hbar}{M_x}\chi^2(d)\left(\frac{\mu_x}{M_x}\right)^2 \times \left[\frac{1}{2k_B}\frac{\partial E_{kin}}{\partial T_0} + 2\frac{E_{kin}}{k_B T_0}\right], \tag{6}$$

where the average thermal energy of QW excitons is given by

$$E_{kin} = k_B \frac{T^2}{T_0}\int_0^\infty \frac{z dz}{\exp[-\mu/(k_B T) + z] - 1} \tag{7}$$

with μ and T_0 defined by Eq. (1). In Fig. 1 we plot $D_x^{(2d)} = D_x^{(2d)}(T, T_0)$ calculated for various T and T_0 (note that T_0 is propotional to n_{2d}) by using Eqs. (6)-(7) with $\chi = a_{2d}^{B}/(2d)$ and $C = 4/\pi$. In the limit of classical, Maxwell-Boltzmann statistics of QW excitons, when $T \gg T_0$, Eqs. (6)-(7) reduce to

$$D_x^{(2d,cl)} = 2C\frac{\hbar}{M_x}\chi^2(d)\left(\frac{\mu_x}{M_x}\right)^2\frac{T}{T_0}\,. \tag{8}$$

Note that for a classical 3D gas of excitons one has $D_x^{(3d)} \propto T^{1/2}$. In the limit of well-developed quantum statistics, $T \ll T_0$, Eqs. (6)-(7) yield

$$D_x^{(2d,qm)} = \frac{\pi^2}{4}C\frac{\hbar}{M_x}\chi^2(d)\left(\frac{\mu_x}{M_x}\right)^2\left(\frac{T}{T_0}\right)^2\,. \tag{9}$$

Drastic reduction of $D_x^{(2d,qm)}$ in comparison with $D_x^{(2d,cl)}$ is due to non-classical accumulation of low-energy QW excitons which occurs at $T < T_0$.

Substitution of Eq. (3) into the expression for $D_T^{(2d)}$ [see Eq. (2)] yields $D_T^{(2d)} = 0$, because $l_{2d}(\partial v_{2d}/\partial T) = v_{2d}(\partial l_{2d}/\partial T)$. This is in a sharp contrast with the 3D case, when $(\partial l_{3d}/\partial T) \simeq 0$ and $\partial v_{3d}/\partial T \propto T^{-1/2}$ give rise to the thermal diffusion coefficient $D_T^{(3d)} \propto T^{1/2}$ for Maxwell-Boltzmann statistics. Thus self-thermodiffusion of QW excitons is negligible in comparison with self-diffusion which is due to the concentration gradient.

The drift flux density of QW excitons, $\mathbf{J}_x^{drift}$, is given by

$$\mathbf{J}_x^{drift} = -\mu^{(2d)} n_{2d} \nabla\left(u_0 n_{2d} + U_{QW}\right)\,, \tag{10}$$

where $\mu^{(2d)}$ is the mobility of quasi-2D excitons and U_{QW} is the in-plane QW potential. The first term in the square brackets on the right hand side of Eq. (10) describes the drift flux due to repulsive exciton-exciton interaction. The exciton mobility is determined through $D_x^{(2d)}$ by

$$\mu^{(2d)} = \frac{D_x^{(2d)}}{k_B T_0}\left[e^{T_0/T} - 1\right]\,. \tag{11}$$

For $T \gg T_0$ Eq. (11) reduces to the Einstein relation, i.e., $\mu^{(2d)} = D_x^{(2d)}/(k_B T)$. In the quantum limit $T \leq T_0$, however, Eq. (11) gives strong n_{2d}-dependent increase of the exciton mobility calculated in terms of $D_x^{(2d)}$.

By using Eqs. (2), (10) and (11), we end up with the nonlinear quantum diffusion equation:

$$\frac{\partial n_{2d}}{\partial t} = \nabla\Big[D_x^{(2d)}\nabla n_{2d} + \frac{2}{\pi}\left(\frac{M_x}{\hbar^2}\right)D_x^{(2d)}\left(e^{T_0/T} - 1\right) \times \nabla\left(u_0 n_{2d} + U_{QW}\right)\Big] - \frac{n_{2d}}{\tau_{opt}} + \Lambda\,, \tag{12}$$

where the optical lifetime $\tau_{opt} = \tau_{opt}(T, T_0)$ of QW excitons is given by Eqs. (32)-(35) of [7], the self-diffusion coefficient $D_x^{(2d)} = D_x^{(2d)}(T, T_0)$ is determined by Eqs. (6)-(7) and $\Lambda = \Lambda(\mathbf{r}_\parallel, t)$ is the generation rate of excitons. Note that the numerical constant $C \simeq 1$ in the expression (6) for $D_x^{(2d)}$ can be calculated by analysing the relevant linearized quantum kinetic equation with standard methods [11,12] developed for classical bulk gases.

III. SCREENING OF LONG-RANGE-CORRELATED DISORDER

One of the most interesting results of the diffusion Eq. (12) is effective screening of long-range-correlated QW disorder by dipole-dipole interaction of indirect excitons. In sharp contrast with excitons in single QWs, the blue-shift of the indirect exciton line, which has already been observed in many experiments (see, e.g., [1,5]), accurately fits the mean-field-theory result, $\delta U = u_0 n_{2d}$, over a broad range of concentrations, $10^8\,\mathrm{cm}^{-2} \leq n_{2d} \leq 10^{11}\,\mathrm{cm}^{-2}$. Here u_0 is given by Eq. (4) with $\chi(d) = a_{2d}^{B}/(2d)$.

In order to demonstrate and estimate analytically the screening effect, we put $1/\tau_{opt} = 0$ (no optical decay) and $\Lambda = 0$ (no source of indirect excitons). In this case a steady-state solution of Eq. (12) can be found analytically. The input, unscreened in-plane random potential $U_{QW} = U_{rand}(\mathbf{r}_\parallel)$ is due to the QW thickness and alloy fluctuations, $\mathbf{r}_\parallel$ is the in-plane coordinate. For average concentrations of indirect QW excitons such that $u_0 n_{2d}^{(0)} \gg |U_{rand}(\mathbf{r}_\parallel)|$ the steady-state solution yields

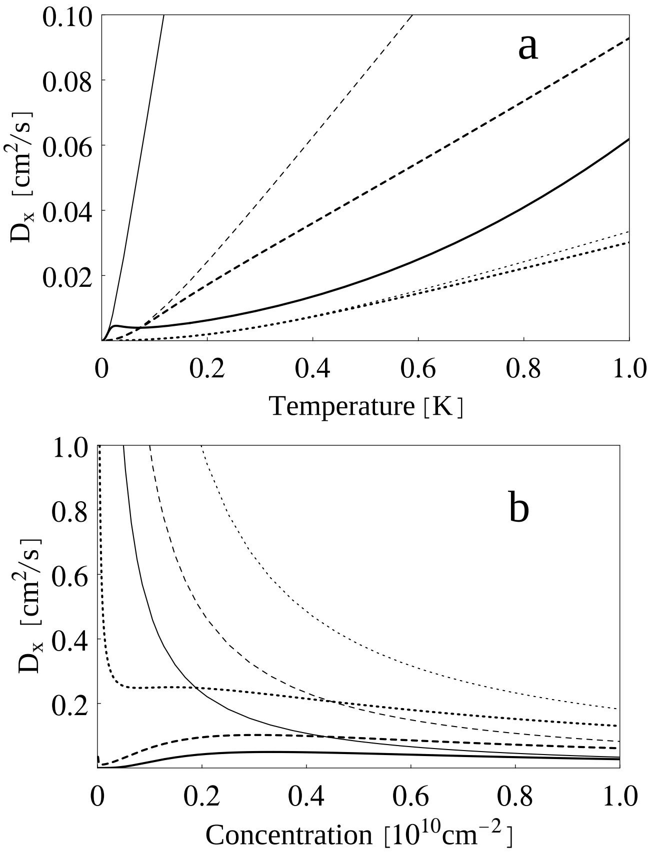

FIG. 1. Diffusion coefficient of indirect excitons in GaAs/AlGaAs coupled QWs calculated using Eqs. (6)-(7) with $C = 4/\pi$ and $\chi = a^{\rm B}_{\rm 2d}/(2d)$. (a) The diffusion coefficient versus T for $n_{\rm 2d} = 10^9\,{\rm cm}^{-2}$ (solid lines), $5 \times 10^9\,{\rm cm}^{-2}$ (dashed lines) and $2 \times 10^{10}\,{\rm cm}^{-2}$ (dotted lines). (b) The diffusion coefficient against $n_{\rm 2d}$ for $T = 0.5\,{\rm K}$ (solid lines), $1\,{\rm K}$ (dashed lines) and $2\,{\rm K}$ (dotted lines). $D_{\rm x}^{(2d)}$ and $\tilde{D}_{\rm x}^{(2d)}$ are shown by the thin and bold lines, respectively. $M_{\rm x} = 0.215\,m_0$, $\mu_{\rm x} = 0.046\,m_0$ and $d = 11.5\,{\rm nm}$.

$$\delta n_{\rm 2d} = - \frac{U_{\rm rand}(\mathbf{r}_\parallel) n^{(0)}_{\rm 2d}}{k_{\rm B}T_0 (e^{T_0/T} - 1)^{-1} + u_0 n^{(0)}_{\rm 2d}} \, ,$$
$$U_{\rm eff} = u_0 n^{(0)}_{\rm 2d} + \frac{U_{\rm rand}(\mathbf{r}_\parallel)}{1 + [(2M_{\rm x})/(\pi\hbar^2)](e^{T_0/T} - 1)u_0} \, , \quad (13)$$

where $\delta n_{\rm 2d} = n_{\rm 2d}(\mathbf{r}_\parallel) - n^{(0)}_{\rm 2d}$. $U_{\rm eff} = U_{\rm rand}(\mathbf{r}_\parallel) + u_0 n_{\rm 2d}(\mathbf{r}_\parallel)$ is the effective, screened in-plane potential. Relaxation of the long-range-correlated random potential is described by the denominator in the expression for $U_{\rm eff}$ [see Eq. (13)]. For $n^{(0)}_{\rm 2d} \gg |U_{\rm rand}/u_0|$ the denominator is much larger than unity. For classical statistics, when $T \gg T_0$, relationships (13) reduce to $\delta n_{\rm 2d}(\mathbf{r}_\parallel) = -[U_{\rm rand}(\mathbf{r}_\parallel) n^{(0)}_{\rm 2d}]/[k_{\rm B}T + u_0 n^{(0)}_{\rm 2d}]$ and $U_{\rm eff}(\mathbf{r}_\parallel) = u_0 n^{(0)}_{\rm 2d} + [U_{\rm rand}(\mathbf{r}_\parallel) k_{\rm B}T]/[k_{\rm B}T + u_0 n^{(0)}_{\rm 2d}]$. For $u_0 n^{(0)}_{\rm 2d} \gg k_{\rm B}T$ the latter expression describes strong suppression of the potential fluctuations, i.e., instead of input $U_{\rm rand}(\mathbf{r}_\parallel)$ one gets $\kappa U_{\rm rand}(\mathbf{r}_\parallel)$ with $\kappa = (k_{\rm B}T)/(u_0 n^{(0)}_{\rm 2d}) \ll 1$. The screening effect becomes particularly strong in the quantum regime, $T_0 \geq T$. Formally this is due to the term $[\exp(T_0/T) - 1]$ in the expressions (13).

In Fig. 2 we plot the effective potential $U_{\rm eff}(x) - u_0 n^{(0)}_{\rm 2d}$ and concentration of excitons, $n_{\rm 2d}(x)$, calculated with Eq. (12) for a model 1D long-range-correlated random potential $U_{\rm rand}(x)$, shown by the bold lines, and realistic values of $n^{(0)}_{\rm 2d}$, D_x, T, $\tau_{\rm opt}$ and amplitude of $U_{\rm rand}$ (about 0.5 meV). Figure 2 clearly illustrates the origin of the screening effect: Accumulation of indirect excitons in the minima of $U_{\rm rand}(\mathbf{r}_\parallel)$ and their depletion at the maxima of $U_{\rm rand}(\mathbf{r}_\parallel)$. In the experiments [1–3] the blueshift of the energy of indirect excitons $\delta U = u_0 n^{(0)}_{\rm 2d} \simeq 1.4 - 1.6\,{\rm meV}$ for $n^{(0)}_{\rm 2d} = 10^{10}\,{\rm cm}^{-2}$. For this moderate concentration of excitons the long-range-correlated in-plane disorder is already drastically screened and relaxed at $T \leq 4.2\,{\rm K}$ (see also Fig. 2). One can directly include the disorder-assisted effects into diffusion Eq. (12) by using a thermionic model. In this case we replace $D^{(2d)}_{\rm x}$ by $\tilde{D}^{(2d)}_{\rm x} = D^{(2d)}_{\rm x} \exp\left[-(U_{\rm eff} - u_0 n^{(0)}_{\rm 2d})/(k_{\rm B}T)\right]$ and remove $U_{\rm QW} = U_{\rm rand}$ from eq. (12). For $T \gg T_0$ the diffusion coefficient $\tilde{D}^{(2d)}_{\rm x}$ is

$$\tilde{D}^{(2d,cl)}_{\rm x} = D^{(2d,cl)}_{\rm x} \exp\left[- \frac{U_{\rm rand}(\mathbf{r}_\parallel)}{k_{\rm B}T + u_0 n^{(0)}_{\rm 2d}} \right] , \qquad (14)$$

where $D^{(2d,cl)}_{\rm x}$ is given by Eq. (8) and $U_{\rm rand}(\mathbf{r}_\parallel)$ can be replaced by $U^{(0)}_{\rm rand} = 2 < |U_{\rm rand}(\mathbf{r}_\parallel)| >$. The calculated diffusion coefficent $\tilde{D}^{(2d)}_{\rm x}$, which is relevant to the analysis of the experimental data [1–3], is shown in Fig. 1. Note that due to the relatively strong dipole-dipole interaction only a dip, natural or externally applied, in-plane potential $U_{\rm trap} \sim 10\,{\rm meV}$ can spatially confine the indirect excitons of high concentrations $n^{(0)}_{\rm 2d} \sim 5 \times 10^{10}\,{\rm cm}^{-2}$ [3,4]. Quantum diffusion of indirect excitons towards a μm-scale trap and their real-space distribution $n_{\rm 2d} = n_{\rm 2d}(\mathbf{r}_\parallel)$ in the trap can be calculated by using Eq. (12) with $\tilde{D}^{(2d)}_{\rm x}$ and $U_{\rm QW} = U_{\rm trap}(\mathbf{r}_\parallel)$ [3].

IV. DISCUSSION

In the experiments [1–3] with GaAs/AlGaAs coupled QWs the effective temperature T of indirect excitons can considerably exceed the bath, cryostat temperature $T_{\rm b}$. The excitons cool down due to interaction with bulk LA-phonons, while incoming, optically-generated high-energy excitons tend to increase T. Thus in order to adapt the quantum diffusion picture to the optical experiments, Eq. (12) should be completed with Eq. (11) of [7]. The latter describes the local temperature $T = T(\mathbf{r}_\parallel, t)$ which is $n_{\rm 2d}$-dependent especially for well-developed BE statistics of QW excitons.

The diffusion transport occurs when $l_{\rm 2d}/a^{(0)}_{\rm 2d} \gg 1$ and when the exciton-exciton interaction is the dominant scattering mechanism. The above conditions limit the

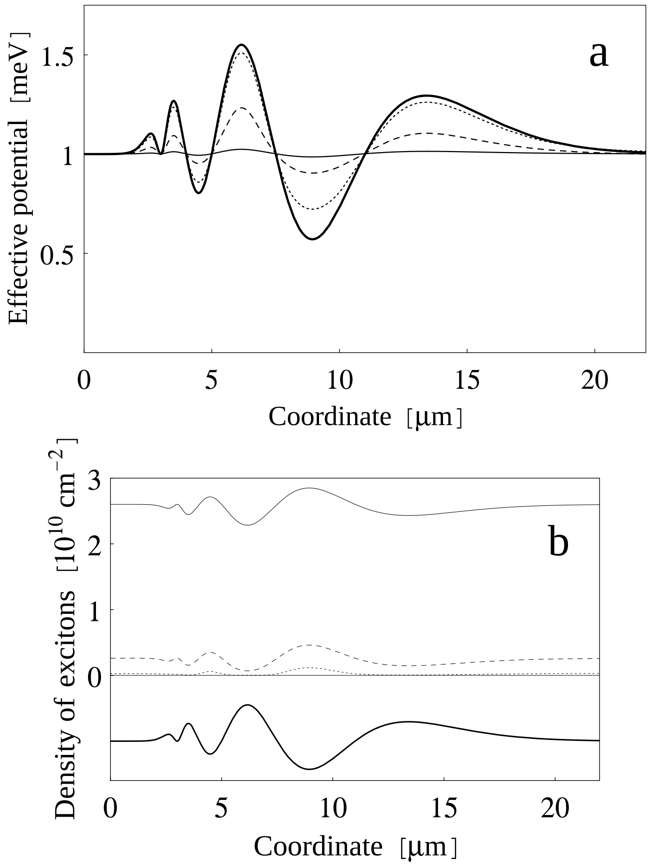

FIG. 2. Relaxation of long-range-correlated disorder by the dipole-dipole interaction of indirect excitons in GaAs/AlGaAs coupled QWs. (a) The effective, screened potential $U_{\rm eff}(x) - u_0 n_{\rm 2d}^{(0)}$ and (b) local concentrations of indirect excitons $n_{\rm 2d}(x)$ versus in-plane coordinate x. The average concentrations are $n_{\rm 2d}^{(0)} = 2.6 \times 10^{10}\,{\rm cm}^{-2}$ (thin solid line), $2.6 \times 10^{9}\,{\rm cm}^{-2}$ (dashed line), and $2.6 \times 10^{8}\,{\rm cm}^{-2}$ (dotted line). Temperature $T = 2\,{\rm K}$, diffusion coefficient $D_x = 100\,{\rm cm}^2/{\rm s}$, and radiative lifetime $\tau_{\rm opt} = 20\,{\rm ns}$. In both figures the input, unscreened potential $U_{\rm rand}(x)$ is shown by bold solid lines.

application of the theory to $10^9\,{\rm cm}^{-2} \leq n_{\rm 2d} \leq 2 \times 10^{10}\,{\rm cm}^{-2}$ for GaAs-based QWs. The developed model is definitely not applicable for $T \leq T_{\rm c}$, where $T_{\rm c}$ is the transition temperature to a superfluid phase. Note that for a very dilute gas of quasi-2D excitons $T_{\rm c} \ll T_0$ [13,14]. Furthermore, in experiments with spatially-inhomogeneous optical generation of QW excitons the diffusive propagation always preceds a possible transition to the superfluid motion of excitons.

By analysing a relevant Gross-Pitaevskii equation we have also found the screening effect for mid-range-correlated (length scale of a few $a_{\rm 2d}^{\rm B}$) in-plane disorder. Namely the dipole-dipole interaction between excitons considerably decreases the localization energy of an indirect exciton in a mesoscopic in-plane trap. This leads to relaxation of the mid-range-correlated QW potential fluctuations. Short-range 2D disorder, which can be described in terms of a random contact potential, is also strongly suppressed by interacting bosons [15]. Thus we conclude that in high-quality GaAs/AlGaAs coupled QWs the in-plane random potential of any correlation length is effectively screened and practically removed at $n_{\rm 2d}^{(0)} \geq 10^{10}\,{\rm cm}^{-2}$, due to the dipole-dipole interaction of indirect excitons. In this case the in-plane momentum of indirect excitons, $\mathbf{p}_\parallel$, becomes a good quantum number even for low-energy particles, as was demonstrated in the experiments [2,16]. Thus the phonon-assisted relaxation and PL kinetics [7–9], formulated in terms of well-defined $\mathbf{p}_\parallel$, are adequate for explanation and modelling of the recent experiments on statistically-degenerate excitons in GaAs/AlGaAs coupled QWs [2].

We attribute a relatively large width $\sim 0.5 - 1\,{\rm meV}$ of the PL line associated with indirect excitons at $n_{\rm 2d}^{(0)} \geq 10^{10}\,{\rm cm}^{-2}$ to intrinsic, homogeneous broadening rather than to a disorder-assisted inhomogeneous width. While the dipole-dipole interaction of indirect excitons is much weaker and of shorter-range than the $1/r_\parallel$ Coulomb law, it gives rise to a relatively large correlation energy in comparison with the leading mean-field-theory correction $\delta U = u_0 n_{\rm 2d}^{(0)}$. This is because screening of the dipole-dipole interaction by indirect excitons is very weak. In this case the optical decay of an indirect exciton interacting with its nearest neighbouring exciton(s) gets an energy uncertainty. The latter depends upon the distance between the interacting particles and results in a large ($\sim 1\,{\rm meV}$) T- and $n_{\rm 2d}$-dependent homogeneous width of indirect excitons in GaAs/AlGaAs coupled QWs.

V. CONCLUSIONS

In this work we have developed a quantum diffusion theory for (indirect) excitons in (coupled) quantum wells. The main results are (i) the quantum diffusion Eq. (12) with the diffusion coefficient, which is calculated within a microscopic picture and is given by Eqs. (6)-(7) and (14), (ii) the modified Einstein relation (11) between the mobility and diffusion coefficient of QW excitons, and (iii) effective screening of QW disorder by dipole-dipole interacting indirect excitons.

ACKNOWLEDGMENTS

The author appreciates valuable discussions with L. V. Butov, D. S. Chemla and L. V. Keldysh. Support of this work by the EPSRC is gratefully acknowledged.

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